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Contribution of Bound States to the Harmonic Generation in Hydrogen at Moderate Laser Intensities

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ABSTRACT

Zero-field parabolic states of hydrogen are used as a basis set for the study of low-order harmonic generation in moderately strong laser fields. The disappearance of bound parabolic states with large electric dipole moments in moderately strong fields leads to the simplification of the expression for the total time-dependent dipole moment of the atom. By including only contributions from bound atomic states and with some simplifying assumptions it is possible to express the total dipole moment by an analytic formula. The present results are compared to the benchmark calculations [1,2].

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I. INTRODUCTION

The interaction of an intense ultrashort pulse laser with an atom, molecule, cluster, or solid target gives rise to a number of competing processes including the generation of harmonic radiation [3]. The major focus on harmonic generation has been on the successful generation of higher order harmonics due to the interest in developing shorter wavelength tunable coherent radiation sources in the VUV- and X-ray regimes of the spectrum for applications in, for example, plasma diagnostics, lithography, and x-ray holography. The spectral distribution of the harmonic intensity as a function of harmonic number exhibits a characteristic spectra displaying a rapid decay of intensity for low harmonics followed by a plateau region where the intensity is relatively insensitive to harmonic number and remains fairly flat only finally to fall precipitously for higher harmonics. The shape of the spectra for the plateau and cutoff harmonics can be reasonably explained in terms of a semiclassical recollision model often referred to as the tunneling-recombination model that predicts a cutoff energy of $I_p + 3.17U_p$ where I_p is the ionization energy and U_p is the ponderomotive energy of the laser field. Once the electron is liberated from the core it is accelerated in the field until the phase of the field reverses and pushes the electron back toward an ion where it may radiatively recombine leading to harmonic generation [4,5]. The semiclassical model has been useful for explaining some of the experimentally observed features. Since its inception the semiclassical model has become the standard with which a number of classical, quantum, and quantum field theoretic models have been compared. It is not our intent to discuss the pros and cons of each model and therefore refer the interested reader to three excellent review articles [6-9] where they compare, contrast, and consider the advantages and disadvantages of each approach. The most recent quantum field model appears in [10-13] and contain adequate references to earlier works. However, by their very nature many of these models are very complicated and computer intensive and although they provide insight and explanations for the purely quantum effects they lose the simplicity characteristic of the semiclassical model. Therefore, in the spirit of the semiclassical model we describe what we believe to be a simple theory that includes some of the effects of

the quantum and classical models but remains physically transparent and calculationally straightforward.

The physics responsible for harmonic generation is due to the asymmetry in the distribution of the electronic charge density in each atom composing the medium by the time-varying driving laser field. The action of the field produces a time-dependent dipole moment whose phase is related to the incident applied laser field and whose Fourier transform is related to the strengths and phases of the harmonic radiation from the individual atoms.

It was not known initially if the harmonics were dominated by single atom interactions or if propagation through the medium controlled the spectral behavior. Actually, single atom calculations, even simple two-level atom approaches, have reproduced many of the features associated with strongly driven nonlinear systems. The characteristic profile of harmonic order as a function of dipole moment, including the plateau and cutoff, is often explained in terms of field induced ionization that becomes the source term for electrons born with zero initial velocity in the laser focus. Following liberation from the atom some of the electrons follow trajectories on the laser half-cycle that return to recollide and radiatively recombine with the nucleus producing harmonic radiation. This semiclassical picture captures many of the observed features of harmonic generation including the correct scaling of the harmonic spectrum with the intensity of the field. In general, the modeling of strong-field harmonic conversion requires both the evaluation of the single atom response and the solution of the propagation equations for the harmonic fields. The propagation calculation requires knowledge of the single-atom dipole strength (and phase) as a function of the intensity of the driving field, for a large number of intensity points, typically on the order of hundreds. This explains why most calculations have been carried out using simple models for describing harmonic generation. In this investigation we will focus on a simplified approach for the single atom aspects of the problem and refer to the work of Borisov et al. [14] for the propagation issues.

It is also worth mentioning that the efficiency of high harmonic generation is not yet high enough for many of the potential applications mentioned above. The problem is twofold; 1) not enough energy can be stored in the higher harmonics and 2) phase mismatch between the fundamental and the harmonic fields can result in the amplitude of a given harmonic changing

dramatically with phase upwards of 3 orders of magnitude depending on the specific case. A possible resolution of this problem might be accomplished by channeling the emitted radiation into a single harmonic by the process of pulse shaping [15]. Finally, since much of the recent emphasis has been on higher-order harmonics we focus on the low-order harmonics and demonstrate that they can be described adequately by the bound atomic states.

II. MODEL AND CALCULATIONS

There are basically two quantum mechanical approaches to the theoretical study of harmonic generation; the first one is based on the direct numerical solution of the time-dependent Schrödinger equation [1,16-18], the other one on the Floquet formalism [2,19,20]. If accurate results are required, both approaches are computationally demanding. This has led to a number of modified and simplified treatments of harmonic generation with various degrees of complexity. Some procedures, besides being simple, may also offer certain insights into individual physical processes that contribute to the generation of harmonic radiation.

The recollision model of harmonic generation, even in its fully quantum theoretical form [21,22], does not include any participation of excited bound states in the emission of high harmonics and is essentially valid for harmonics with energy well above the ionization energy [21]. On the other hand, several previous studies [23-29] showed that excited levels may be of importance for harmonic generation. In the present paper we investigate the effect of radiation on excited levels and show that the harmonic emission of hydrogen at moderate laser intensities may be explained by transitions between the side-bands of these levels and the ground level.

We study the hydrogen atom in the presence of a moderately intense linearly polarized radiation field of infinite duration and look for an approximate stationary solution of the Schrödinger equation in order to obtain a simple analytic expression describing the harmonic radiation.

The wave function of a hydrogen atom in a linearly polarized radiation field may be conveniently expressed in terms of zero-field parabolic states $|n\ n_1\ m\rangle$. An important property

of these states is the generally asymmetric charge distribution with respect to the xy plane, indicating an increasing possibility of field ionization for some states with nonvanishing values of the dipole moment $\langle n\, n_1\, m\,|z|\,n\, n_1\, m\rangle$. In order to obtain a simple expression for the dipole moment of the atom in the radiation field, including only contributions from bound states, we adopt the following two simplifications: the continuum states of the atom will be ignored and all matrix elements $\langle n\, n_1\, m\,|z|\,n'\, n_1'\, m'\rangle$ taken between excited states with $n \neq n'$ will be omitted. These simplifications allow us to express the total induced dipole moment of the atom in closed form as a function of the field amplitude and a set of atomic parameters. With the electric field in the z-direction, the part of the Hamiltonian representing the interaction between the bound electron and the field in the dipole approximation has the form $F z \sin\omega t$. In our procedure the $|n\ n_1\, m\rangle$ states are coupled only to the ground state and the total wave function Ψ will contain only parabolic states with $m = 0$. In the subsequent equations the label m will be omitted and atomic units will be used throughout the paper unless indicated otherwise.

In the basis of parabolic states, the matrix with elements $\langle n n_1 |z| n n_1\rangle$ is diagonal and

$$\langle n n_1 \,|\, z \,|\, n n_1\rangle = \frac{3}{2} n(n_1 - n_2) \tag{1}$$

with $n_2 = n - n_1 - 1$. Factors $(n_1 - n_2)$ assume values $-(n-1), -(n-3), \ldots n-3, n-1$. For our purpose, it is convenient to designate each parabolic state by $|n\,\lambda\rangle$ according to the value of the matrix element (1) so that

$d_{n\lambda} = \langle n\lambda \,|\, z \,|\, n\,\lambda\rangle = 3n\lambda$ for n odd,

$$= \langle n\lambda \,|\, z \,|\, n\lambda\rangle = 3n(\lambda \pm \frac{1}{2}) \quad \text{for } n \text{ even and } \lambda \lessgtr 0. \tag{2}$$

For n odd, $\lambda = 0, \pm 1, \pm 2, \ldots \pm \frac{1}{2}(n-1)$, and for n even, $\lambda = \pm 1, \pm 2, \cdots; \pm \frac{1}{2} n$. The functions $|n\,\lambda\rangle$ of the zero-field parabolic states are superpositions of regular spherical functions $\varphi_{n\ell}$ represented as

$$|n\ \lambda\rangle = \sum_{\ell} b_{n\ell\lambda} \varphi_{n\ell} \tag{3}$$

and $b_{n\ell\lambda}$ can be expressed in terms of 3-j symbols [30,31]. With the convention that $b_{n\ell\lambda}$ are positive for $\ell=0$, we have

$$b_{n\ell\lambda}=(-1)^{\alpha-(n-1)/2}\ (2\ell+1)^{1/2}\begin{pmatrix}(n-1)/2 & (n-1)/2 & \ell\\ \alpha & -\alpha & 0\end{pmatrix}, \tag{4}$$

where $\alpha=\lambda$ for n odd, $\lambda\pm\frac{1}{2}$ for n even and $\lambda \lessgtr 0$, with special values

$$b_{n0\lambda}=n^{-1/2},\quad b_{n1\lambda}=2\sqrt{3}\alpha[n(n^2-1)]^{-1/2}. \tag{5}$$

Following the Floquet method, we require the total wave function to have the form

$$\Psi=\sum_{n\lambda} B_{n\lambda}(t)\,|n\lambda\rangle\,\exp\left[-i(E_1+\varepsilon)t\right]. \tag{6}$$

E_1 is the energy of the zero-field ground state and $(E_1+\varepsilon)$ is the quasi-energy of the system. Due to the periodicity of the Hamiltonian it is possible to write $B_{n\lambda}(t)$ in the form

$$B_{n\lambda}(t)=C_{n\lambda}(t)\exp(i\,\xi_{n\lambda}\cos\omega t) \tag{7}$$

and the solution of the Schrödinger equation may be obtained in a convenient form if constants $\xi_{n\lambda}$ are properly chosen. With the definition

$$\xi_{n\lambda}=F\omega^{-1}d_{n\lambda} \tag{8}$$

the matrix elements of the operator

$$i\frac{\partial}{\partial t}-(H_0+Fz\sin\omega t),$$

where H_o is the field-free Hamiltonian, are zero in the basis of functions

$$|n\lambda\rangle\exp(i\,\xi_{n\lambda}\cos\omega t-i\,E_n t\rangle$$

for a given n. Therefore the strongest effect of the field on the atom is already properly included in the basis set if $B_{n\lambda}$ in (6) are defined according to (7) and (8). Parabolic states $|n0\rangle$ are not coupled to the ground state $|10\rangle$ due to (5) and they will not be included in the expansion (6). Using the substitution

$$C_{10}=\sum_p i^p C_{10}^{(p)}e^{ip\omega t},\quad C_{n\lambda}=\sum_q i^q C_{n\lambda}^{(q)}e^{iq\omega t} \tag{9}$$

together with the expansion of the exponential factor in (7) in terms of Bessel functions $J_\mu(\xi_{n\lambda})$, one derives a system of coupled equations for $C_{10}^{(p)}$ and $C_{n\lambda}^{(q)}$ (with $n > 1$). We require a solution for which $C_{10}^{(0)} = 1$ in the limit of vanishing field. Then from the properties of $b_{n1\lambda}$ and $\xi_{n\lambda}$ it follows that all $C_{10}^{(p)}$ with p odd should be zero and that the matrix of coefficients for $C_{10}^{(p)}$ and $C_{n\lambda}^{(q)}$ is real and symmetric. Moreover we find that

$$C_{n\lambda}^{(q)} = (-1)^q C_{n,-\lambda}^{(q)} \ . \tag{10}$$

The sum over two terms with opposite values of λ may be readily evaluated and the system may be further reduced to the positive values of λ.

With $\lambda > 0$ and p even, we obtain

$$\begin{aligned} &(\varepsilon - p\omega) C_{10}^{(p)} + 2 \sum_{n\lambda q} R_{n\lambda pq} C_{n\lambda}^{(q)} = 0, \\ &(\varepsilon + E_1 - E_n - q\omega) C_{n\lambda}^{(q)} + \sum_p R_{n\lambda pq} C_{10}^{(p)} = 0, \\ &R_{n\lambda pq} = F\, a(10, n1)\, b_{n1\lambda}(p-q)\, \xi_{n\lambda}^{-1} J_{p-q}(\xi_{n\lambda}). \end{aligned} \tag{11}$$

$a(n\ell, n'\ell')$ is a matrix element $< \varphi_{n\ell} | z | \varphi_{n'\ell'} >$ taken between spherical states with m=0, and E_n are zero field energies of the $n\lambda$ states. The summation over p and q is from $-\infty$ to $+\infty$ and the summation over $n\lambda$ includes all parabolic states $|n\lambda\rangle$ for $n > 1$ and $\lambda > 0$ which may exist at a particular amplitude F of the electric field. Conditions for the inclusion of $|n\lambda\rangle$ states to the sum in (11) will be discussed later in par. III. The solution of (11) yields relative values of coefficients $C_{10}^{(p)}, C_{n\lambda}^{(q)}$ and eigenvalues ε.

To obtain the dipole moment of the atom in the radiation field, we chose of all possible solutions of (11) the one with the largest value of $C_{10}^{(0)}$, which has a character of the unperturbed ground state. Such a solution corresponds to the smallest absolute value of ε, at least for small values of the field amplitude F. The time-dependent dipole moment in the length gauge is given by

$$D(t) = \langle \Psi | z | \Psi \rangle \tag{12}$$

and it may be written as a sum over terms with time factors $e^{\pm i(q+\mu-p)\omega t}$. In evaluating (12) we include only terms with matrix elements connecting the ground level with the excited $n\lambda$ levels which are expected to give dominant contributions to the dipole moment. Matrix elements $\langle 10|z|n\lambda\rangle$ are real and the summation over $\pm\lambda$ in the expression for $D(t)$ leads to cancellation of all terms with factors $e^{\pm i(q+\mu-p)\omega t}$ for which $q+\mu-p$ is even. We define an odd integer

$$N = |q+\mu-p| \tag{13}$$

and obtain

$$D(t) = \sum_N \left(D_N e^{iN\omega t} + c.c. \right) \tag{14}$$

with

$$D_N = 2 \sum_{n\lambda pq\mu} a(10, n1)\, b_{n1\lambda} i^N C_{10}^{(p)} C_{n\lambda}^{(q)} J_\mu(\xi_{n\lambda}). \tag{15}$$

As in (11), the summation over parabolic states $|n\lambda\rangle$ in (15) is limited due to the presence of the electric field, p is even, $\lambda > 0$ and p, q, μ may have values from $-\infty$ to $+\infty$ and should satisfy the condition (13). Amplitudes $C_{10}^{(p)}$ and $C_{n\lambda}^{(q)}$ in (15) should be normalized. The rate of emission of harmonic photons with energy $\omega' = N\omega$ by an isolated atom is then

$$\Gamma_N = \frac{4}{3}\left(\frac{N\omega}{c}\right)^3 |D_N|^2. \tag{16}$$

III. EFFECTS OF IONIZATION

The simplicity of expression (15) for D_N is achieved mainly because of the omission of continuum states in expansion (6) for the total wave function and to factorization of $B_{n\lambda}$ in (7). An important consequence of our simplifying assumption is the neglect of all processes leading

to ionization. Field ionization of excited $n\lambda$ levels will decrease the availability of these levels for the generation of harmonic emission. Maintaining the simplicity of the expression for D_N, one may approximately take into account the effects of ionization due to the electric field by multiplying all contributions from $n\lambda$ levels to the dipole moment (15) by a factor representing the expectation value $P_{n\lambda}$ of these levels in the AC field. To determine $P_{n\lambda}$ for a given amplitude of the field, we will assume first of all that $P_{n\lambda}$ is zero if F is substantially larger than a certain critical amplitude F_c. In the static field, the threshold value of the field strength necessary for ionization of the n-level is $(2n)^{-4}$. However, for parabolic states F_c will depend on λ within the same n-manifold due to the asymmetry of the charge distribution with respect to the xy-plane. This effect has been studied for static fields by Banks and Leopold [32], Harmin [33], and Popov, et. al. [34]. A better approximation applicable to parabolic states with largest tunneling ionization rate for a given n and field in the $+z$ direction, i.e. with

$\lambda = -\frac{1}{2}n$ for n even and

$\lambda = -\frac{1}{2}(n-1)$ for n odd, yields

$$F_c = 0.130n^{-4} \qquad \text{(Krainov, et. al. [35])} \qquad (17)$$

For such field strength the distance of the saddle point of the potential from the nucleus along the z-axis is equal to $r_s = F_c^{-1/2} \approx 2.77n^2$. This expression is similar to the expression for $|d_{n\lambda}|$ of these levels, namely $|d_{n\lambda}| = \frac{3}{2}n^2$ for n even and $\frac{3}{2}n(n-1)$ for n odd. The similarity becomes even more apparent if we consider that (17) represents only the lower limit of the correct value of F_c [36]. We may therefore approximately find the value of the critical field by setting

$$F_c \approx d_{n\lambda}^{-2}. \qquad (18)$$

According to eq. (18) the center of gravity of the electron mass coincides with the saddle point of the potential. This is in agreement with the fact that ionization can really occur just in or near the region where the Coulomb field is smaller than the external field, and therefore we may generalize the validity of (18) to all $\lambda < 0$ states of the same n-manifold and to field strengths

even larger than $(2n)^{-4}$. Eq. (18) is consistent with the strong λ – dependence of ionization rates found by Luc-Koenig and Bachelier [37] in a static electric field.

In the AC field, the λ – dependence of F_c is due to the splitting of the n-levels into the $n\ell$ or equivalently to $n\lambda$ components (Delone and Krainov [38]) and to overlap of these components with adjacent $n \pm 1$ levels. Assuming the validity of (18) also for the oscillating fields and for levels with $\lambda > 0$, then for the most weakly bound parabolic states with a given n, F_c behaves as n^{-4}, and for the most tightly bound states (with smallest non-vanishing $|d_{n\lambda}|$), F_c scales as n^{-2} in the limit of large n.

Our calculations were performed for radiation intensities from 10^{12} to 8×10^{13} W/cm^2 for the wavelength 1064 nm (ω = 0.04282). The lowest intensity corresponds to F =0.005345 and to $|d_{n\lambda}|$=13.68 according to (18). At this intensity only the following excited $n\lambda$ levels satisfy the condition $|d_{n\lambda}|$<13.68:

n	$\|\lambda\|$	$\|d_{n\lambda}\|$
2	1	3
3	1	9
4	1	6
6	1	9
8	1	12

We note that only levels with the smallest value of $|d_{n\lambda}|$ for each n are present above F =0.005345 and thus the summation over $n\lambda$ in (15) reduces to a very small number of terms.

At the intensity of 8×10^{13} W/cm^2 only the lowest excited state with n = 2 could be present according to the condition (18). In reality the expectation value $P_{n\lambda}$ continuously changes in a broad range around F_c and it depends on the actual process of ionization and therefore on the Keldysh parameter γ. This parameter varies from 4.0 to 0.11 from n = 2 to n = 8 in the entire interval of intensities. Consequently, no unique process will be solely responsible for destruction of bound states at all intensities. For simplicity we have adopted the

photoionization as the dominant process and determined $P_{n\lambda}$ for all considered intensities from the expression

$$P_{n\lambda}=(1+2\pi n^3 W_{n\lambda})^{-1} \tag{19}$$

where $2\pi n^3$ is the Kepler orbital period for the n th level and $W_{n\lambda}$ is the multiphoton ionization rate of the $n\lambda$ state. Because of the approximate character of the expression (18) for the critical field F_c, we have also included levels with n=5 ($|d_{n\lambda}|$=15).

Multiphoton ionization rates W were calculated using a Keldysh-like method in which the final state is represented by a Coulomb-Volkov continuum. This method was proposed by Pan [39] and later used in ref. [40], where a detailed description is given. Because of the value of ω, levels with n=2 and 3 need more than one photon to be ionized. Table 1 shows values of $P_{n\lambda}$ for all included levels and intensities of radiation.

Table 1.

Expectation values $P_{n\lambda}$ of parabolic states $n,|\lambda|=1$.

n / I(10^{13} W/cm^2)	2	3	4	5	6	8
0.1	0.972	0.958	0.620	0.671	0.761	0.728
0.5	0.628	0.482	0.243	0.322	0.391	0.359
1	0.411	0.212	0.138	0.180	0.248	0.233
2	0.357	0.112	0.079	0.125	0.158	0.161
4	0.314	0.157	0.054	0.115	0.121	0.133
8	0.274	0.206	0.044	0.105	0.109	0.129

We note that $P_{n\lambda}$ decreases more rapidly for n=2,3 than for other levels, obviously as a consequence of non-existent single photon ionization for these levels. The decrease of $P_{n\lambda}$ for

all levels becomes slower at intensities above 10^{13} W/cm^2, and the n=3 level shows an indication of a possible onset of stabilization.

IV. RESULTS AND DISCUSSION

The results of our calculation for hydrogen according to (15) with the inclusion of factors $P_{n\lambda}$ from Table 1 are shown on Figs. 1 and 2 for wavelength = 1064 nm ($\omega = 0.04282$) and I = 10^{12} to $8\text{x}10^{13}$ W/cm^2. The harmonic spectra on the Figures are compared to grid calculations of Krause et. al. [1] for I > 10^{13} W/cm^2 and to results of the Floquet method by Potvliege and Shakeshaft [2] for I $\leq$ 10^{13}. Relative data for emission rates in [2] were first converted to $|D_N|^2$ according to (16) and then to absolute values using values of $|D_3|^2$ calculated by Krause et. al. [1]. This appears well justified due to excellent agreement between relative values of both results. Data for N = 1 were taken from [2] for all values of I.

Under certain conditions expression (15) for D_N may be further simplified. For low values of F the coefficients $C_{10}^{(p)}$ for $p \neq 0$ are very small and the eigenvalue ε approaches zero. In our calculation for intensities up to $8\text{x}10^{13}$ W/cm^2 for hydrogen we observed that the contribution of terms with $p \neq 0$ were small and that |ε| was always smaller than 0.05. Moreover, if ω is such that $|E_1 - E_n - q\omega|$ is not close to zero for any n and q, it is possible to find an approximate expression for $C_{n\lambda}^{(q)}$ from the second equation in (11) by setting $C_{10}^{(p)} = \delta(p,0), \varepsilon = 0$. Substitution into (15) and inclusion of factors $P_{n\lambda}$ leads to a closed form for D_N,

$$D_N = 2i^N F \sum_{n\lambda q} [a(10,n1)b_{n1\lambda}]^2 (E_1 - E_n - q\omega)^{-1} q\xi_{n\lambda}^{-1} J_{-q}(\xi_{n\lambda})[J_{N-q}(\xi_{n\lambda}) + J_{-N-q}(\xi_{n\lambda})]P_{n\lambda}. \quad (20)$$

The summation over $n\lambda$ and q is the same as in (11). The harmonic spectrum of hydrogen according to (20) agrees in most cases within a factor of 2 with results presented on Figs. 1 and 2. The difference decreases with increasing N and with decreasing intensity of radiation.

The basic features of harmonic spectra are well reproduced in the present calculations: steep decline of $|D_N|^2$ for low harmonics, the onset of a plateau in the vicinity of N=7 and then a rapid decrease. This behavior and the value of the plateau may be explained by the properties of

Bessel functions as discussed in detail by Burlon et al. [27] in the case of a degenerate two-level model atom.

It has been shown by Krause et. al. [1] that $|D_N|^2$ of higher harmonics exhibits rapid fluctuations of up to one order of magnitude as a function of the radiation intensity I. We have found similar behavior in our calculations, but various local maxima and minima do not generally agree with [1] so that occasionally large discrepancies may occur for a particular value of I or N, e.g. for I = 4x10^{13} W/cm^2, N = 17 and I = 8x10^{13} W/cm^2, N = 3,19. Nevertheless, some details of the I-dependence of several harmonics reported in [1] are well reproduced in our results, e.g. the broad and flat maximum between I = 10^{13} and 2x10^{13} W/cm^2 of N = 7 and 9, and a smooth behavior of N = 11 and 19 in approximately the same region.

The time-dependent dipole moment of the atom in the velocity gauge is defined, instead of (12), by

$$D^V(t) = -i\left\langle \Psi \left| \frac{\partial}{\partial z} \right| \Psi \right\rangle = \sum_N (D_N^V e^{iN\omega t} + cc). \qquad (21)$$

Because $\left\langle 10 \left| \frac{\partial}{\partial z} \right| n\lambda \right\rangle = (E_n - E_1)a(10, n1)b_{n1\lambda}$, D_N^V may be obtained from D_N by multiplying each term in (15) by a factor $-i(E_n - E_1)$.

The emission rate Γ_N of harmonic photons with energy $\omega' = N\omega$ is then again given by (16), but D_N should be replaced by $D_N^V(N\omega)^{-1}$. As a result, the emission rate obtained from the velocity expression is close to the rate from the length formula for such harmonics for which the ratio $(E_n - E_1)/N\omega$ of contributing $n\lambda$ levels is close to 1, i.e. for values of N around 11. For lower N, the velocity rate is higher, and for larger N it is lower than the length rate. For example, at 8x10^{13} W/cm^2 the velocity rate is approximately 10 times larger than the length rate for N =3, and 10 times smaller for N =33.

For the intensity of 8x10^{13} W/cm^2 and even greater intensities, higher harmonics require the presence of levels with $|l_{n\lambda}| > 20$, but such levels cannot exist according to our assumption

regarding the critical value F_c. Therefore, the mechanism of harmonic generation in hydrogen described in this paper is questionable beyond 10^{14} W/cm^2, unless a stabilization of high bound levels takes place.

In conclusion, it was shown that with some simplifying assumptions and zero-field bound parabolic states as basis set it is possible to express the time-dependent dipole moment of a hydrogen atom in a radiation field by a simple analytic formula (20). According to the present description, the harmonic radiation of photons with energy up to two times the ionization energy may be interpreted as the result of transitions between the ground state and the energy side-bands of excited states, which are coupled to the ground level by the incident radiation. The number of parabolic states included in the expression for the dipole moment depends on the radiation intensity, and the critical amplitude F_c excluding levels with higher values of $|d_{n\lambda}|$ is given by (18). Consequently, in the interval of radiation intensities beyond 10^{12} W cm^{-2}, only the parabolic levels with $|\lambda| = 1$ may exist and F_c scales as n^{-2}. According to the present results, almost all harmonics in the investigated intensity range may be explained just by the contributions from bound states. Our assumptions restrict the applicability of this procedure to radiation intensities less than 10^{14} W/cm^2 for hydrogen.

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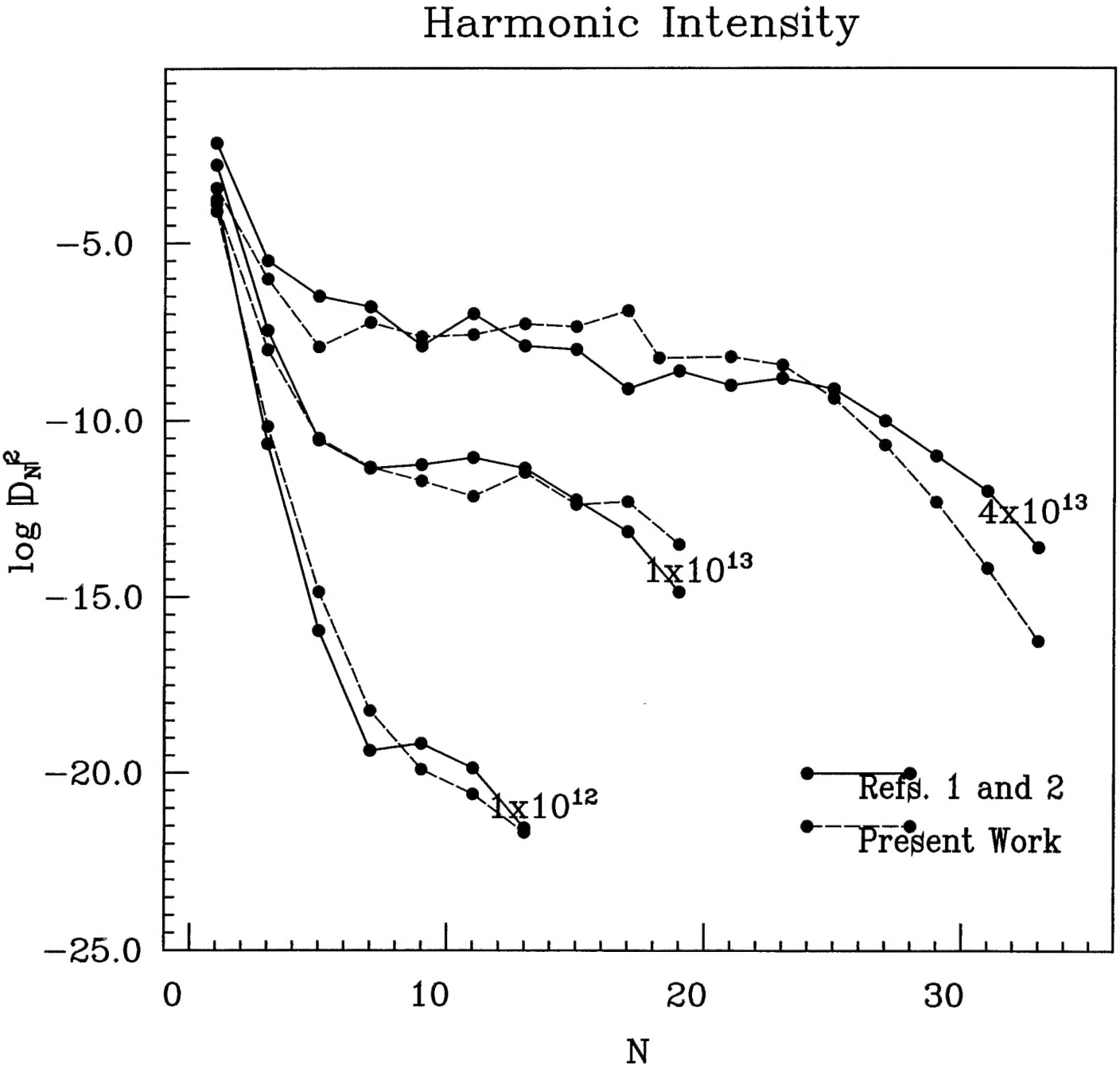
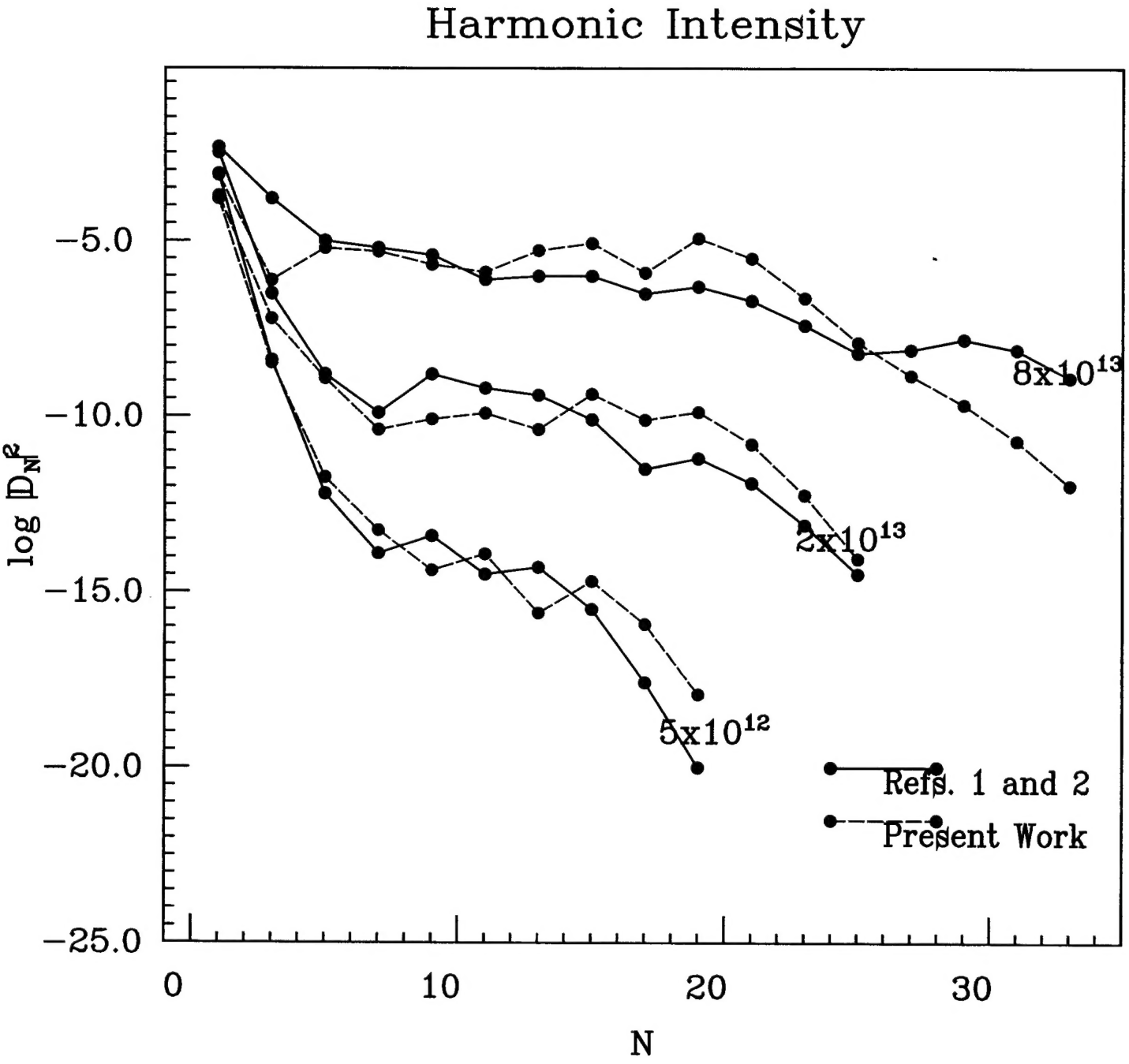
CAPTIONS TO FIGURES

Figure 1. Harmonic intensities $|D_N|^2$ of hydrogen atom at 1064 nm for various laser intensities (W/cm^2), compared with calculations of Krause, et al[1] and Potvliege and Shakeshaft [2].

Figure 2. Same as Fig. 1.


Harmonic Intensity
log D_N^2
−5.0
−10.0
−15.0
−20.0
−25.0
0
10
20
30
N
4x10^13
1x10^13
1x10^12
Refs. 1 and 2
Present Work


Harmonic Intensity
−5.0
−10.0
−15.0
−20.0
−25.0
log D_N^2
0
10
20
30
N
8x10^13
2x10^13
5x10^12
Refs. 1 and 2
Present Work